Supporting information:

Plasmon-assisted spin transition in gold nanostar@Spin Crossover heterostructures

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Figure S1. ATR-FTIR spectra of PVP-stabilized AuNSs before and after the Htrz addition.



Figure S2. XPS spectra of a) N1s, b) Fe2p, c) F1s and d) Survey for NSs@SCO synthesized under inert atmosphere.



Figure S3. TEM images of the Au nanostars (a), nanorods (b), nanotriangles (c), and Ag nanospheres (d).



Figure S4. Characterization of the NS@SCO heterostructure synthesized in air.



Figure S5. TEM of the NS@SCO heterostructure showing how the NP size is estimated.

 Table S1.
 Summary of Fe 2p XPS peak position.

	Fe2p _{3/2}	Fe2p _{3/2} satellite	Fe2p _{1/2}	Fe2p _{1/2} satellite
Fe ²⁺	708.7 eV	-	721.1 eV	-
Fe ³⁺	710.6 eV	715.6 eV	723.3 eV	730.6 eV



Figure S6. Deconvoluted Fe2p region of the NS@SCO synthesized in air (a), and NS@SCO synthesized under inert atmosphere but after 1 day of O₂ ambient exposure (b).



Figure S7. Synthesis performed without adding triazole in the first step.



Figure S8. TEM images different core@shell with a SCO shell and Au nanostars (top-left), nanotriangles (top-right), nanorods (bottom-left), and Ag nanospheres (bottom-right) as the core.



Figure S9. TEM image of the Au nanorods@SCO (a), and EDX mapping of the same area (b). Green and red dots correspond to Fe and Au. The scale bar of (b) is the same as (a).



Figure S10. TEM image of the Au nanotriangles@SCO (a), and EDX mapping of the same area (b). Green and red dots correspond to Fe and Au, respectively. The scale bar of (b) is the same as (a).



Figure S11. TEM image of the Ag nanospheres@SCO (a), and EDX mapping of the same area (b). Green and orange dots correspond to Fe and Ag, respectively. The scale bar of (b) is the same as (a).



Figure S12. DLS plot for PVP-stabilized NS and NS@SCO.



Figure S13. Room temperature PXRD for NS@SCO (black) and SCO (red) compounds. Grey points corresponds to the HTS platform signal. This signal shows up when a small amount of material is measured.

	Peak position / °C	Area / mJ
1º Heating	103	2.7
1º Cooling	66	2.5
2º Heating	87	2.5
2º Cooling	66	2.5
3º Heating	84	2.4
3º Cooling	65	2.5
4º Heating	91	2.4
4º Cooling	66	2.3
0.15 mW·cm ⁻² irradiation Heating	87	1.1
0.15 mW·cm ⁻² irradiation Cooling	65	2.3
0.30 mW·cm ⁻² irradiation Heating	84	0.9
0.30 mW·cm ⁻² irradiation Cooling	65	2.
10 min at 70ºC heating	95	1.9
10 min at 70ºC cooling	67	2.0

Summary of the DSC peaks and area under the curve of the NS@SCO.



Figure S14. Evolution of the areas under the curve extracted from the DSC measurements of the NS@SCO for the first 3 cycles.

 Table S3.
 Summary of the DSC peaks and area under the curve of pure SCO.

	Peak position / °C	Area ∕ mJ·mg⁻¹
1º Heating	114	49
1º Cooling	78	45
2º Heating	109	44
2º Cooling	77	44
0.30 mW·cm⁻² irradiation Heating	111	42
0.30 mW·cm ⁻² irradiation Cooling	76	45



Figure S15. DSC of the SCO in dark (two cycles) and after irradiation with an 808 nm laser at 85 $^{\circ}$ C for 10 min and a power of 0.30 mW·cm⁻².



Figure S16. TEM image of bare SCO NPs.



Figure S17. IV characteristics of the NS@SCO NPs recorded at different temperatures during the heating and the cooling. The inset shows the short-circuit recorded after cooling down to 30 °C.